Life cycle carbon efficiency of Direct Air Capture systems with strong hydroxide sorbents

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Summary

Direct Air Capture (DAC) of carbon dioxide (CO_2) from ambient air has the potential to combat climate change. DAC systems capture CO_2 using a sorbent material and compress it for storage. In this study, we calculated the life cycle carbon efficiency (E_c) of a DAC system which equals the net amount of carbon stored per amount of carbon captured from capture to geological storage. We included greenhouse gas (GHG) emissions during construction of the necessary facilities as well as GHG emissions from energy, water and chemicals needed during operations. The system we analysed includes a hydroxide solution as sorbent material and utilizes the pelletized variant of the Kraft process to regenerate the sorbent and separate the CO_2. To our knowledge, the potential climate benefit over the full life cycle of this DAC system has not been fully investigated up to now. Using the baseline scenario, we obtained a positive E_c of 62%. For the optimistic and pessimistic scenarios, we found a E_c of 93% and 10%, respectively. We conclude that this type of DAC system may be a feasible option to help keep limit global temperature increases to well below 2 ˚C.

Keywords: artificial trees, life cycle analysis, carbon balance, CO_2 stabilization, climate change mitigation, DAC

Highlights

- We calculated the life cycle carbon efficiency of a Direct Air Capture (DAC) system.
- The main GHG emissions of the system are from fossil energy requirements.
- DAC, using hydroxide solutions, can help mitigate climate change.

1 Introduction

Atmospheric concentrations of CO_2 have increased by over 40% since pre-industrial times to over 400 ppm (Scripps Institution of Oceanography 2017). To limit temperature increase to well below 2 ˚C by the end of the 21st century, CO_2 concentrations need to stabilize at about 450 ppm. To achieve this stabilization, emission scenarios show that a 40 to 70% reduction in greenhouse gas (GHG) emission by 2050 is necessary and often include a net CO_2 uptake from the atmosphere by 2100 (Peters et al. 2013, Pachauri et al. 2014).

Because fossil fuels are expected to remain the main energy source in the coming decades (International Energy Agency 2016), methods to capture and store CO_2 are considered important to achieve the climate targets. Carbon capture and storage (CCS) at large industrial point source emission sites may help reduce emissions (Metz et al. 2005, Kenarsari et al. 2013, Volkart et al. 2013, Zhang et al. 2014, Leeson et al. 2017). However, industrial sources are currently estimated to be responsible for about 36% of the GHG emissions (IPCC 2014). Other sources of CO_2 emissions are, generally less localized making on site capture difficult. One method that has been proposed to reduce CO_2 concentrations in ambient air is the use of direct air capture (DAC) systems, sometimes called artificial trees (Lackner et al. 2001). Besides dealing with emissions from non-localized sources, these systems can also help reduce CO_2 concentrations to pre-industrial levels (Lackner et al. 2012).

DAC systems capture CO_2 using a sorbent material and release it in high purity compressed CO_2 streams ready for transport and storage. The core of these systems consists of an air collector where ambient air is brought in contact with the sorbent material and a regeneration facility where the captured CO_2 is separated from the saturated sorbent material. Regeneration and circulation of the sorbent material and circulation of ambient air through these systems require substantial energy inputs, raising questions about the economic and environmental feasibility of this technique as a climate change mitigation option (Goeppert et al. 2012).

Several studies have investigated the economic feasibility of DAC systems with reported costs ranging from $30 to $1000 per tonne of CO_2 removed (Lackner 2009, House et al. 2011, Goeppert et al. 2012). With such a wide range of estimated capital costs, there is no consensus on the economic feasibility of DAC. For example, Simon et al. (2011) argued DAC can be a cost-effective contributor to climate mitigation in the future, while Ranjan and Herzog (2011) found that the costs of DAC systems is prohibitively high compared to other mitigation options.

The environmental feasibility of DAC as a climate change mitigation option can be expressed as the life cycle carbon efficiency (E_c) of the system. E_c is defined as the relative net amount of CO_2 captured by the system, taking into account the GHG emissions, expressed in CO_2 equivalents, caused by the construction and operation of the DAC system. So far, environmental studies on DAC focussed on the energy use of the system. Energy requirements of operating systems have been reported between 50 and 700kJ per mole CO_2 captured (Keith et al. 2006, Lackner 2009, House et al. 2011). At the upper range of these energy requirements, the system only effectively captures CO_2 when a renewable energy source is used. The majority of these studies on the energy requirements of DAC systems, however, are simple energy assessments based on thermodynamic principles and reaction enthalpies instead of outdoor prototypes (House et al. 2011, Ranjan and Herzog 2011, Simon et al. 2011, McGlashan et al. 2012, Lackner 2013, Wilcox et al. 2017).These studies often only include the capture and regeneration step of the DAC systems and thus neglect the energy use from CO_2 transportation and storage. Furthermore, by only analysing energy

consumption, emissions due to the production and use of materials and chemicals needed are neglected. So far, no studies on the environmental effectiveness of hydroxide based DAC systems have included the full system from capture to storage nor have been based on outdoor prototypes.

The aim of this study was to assess the life cycle carbon efficiency of DAC systems and to identify the major contributors to the carbon balance. We included the full life cycle of DAC from capture to geological storage and included CO_2 as well as other GHG emissions during construction of the necessary facilities as well as operating GHG emissions from energy and other resources needed. To this end, we collected experimental data and system descriptions from laboratory and prototype DAC systems using strong hydroxide solutions as sorbents. This data was supplemented with data from conventional Carbon Capture and Storage (CCS) plants to estimate emissions from CO_2 compression, transport and geological storage.

2 Methods

2.1 System description

The Direct Air Capture (DAC) system analysed is designed for an annual capture rate of 1 Mt CO_2 per year and includes 10 air contactor units dispersed around a regeneration and compression facility, CO_2 transport pipelines to the geological storage site and an injection facility. In the air contactor, CO_2 is extracted from ambient air by binding to a sorbent. The captured CO_2 is separated from the sorbent into a pure CO_2 stream and the sorbent is regenerated in the regeneration facility. A schematic representation of the different components of the system is given in figure 1.

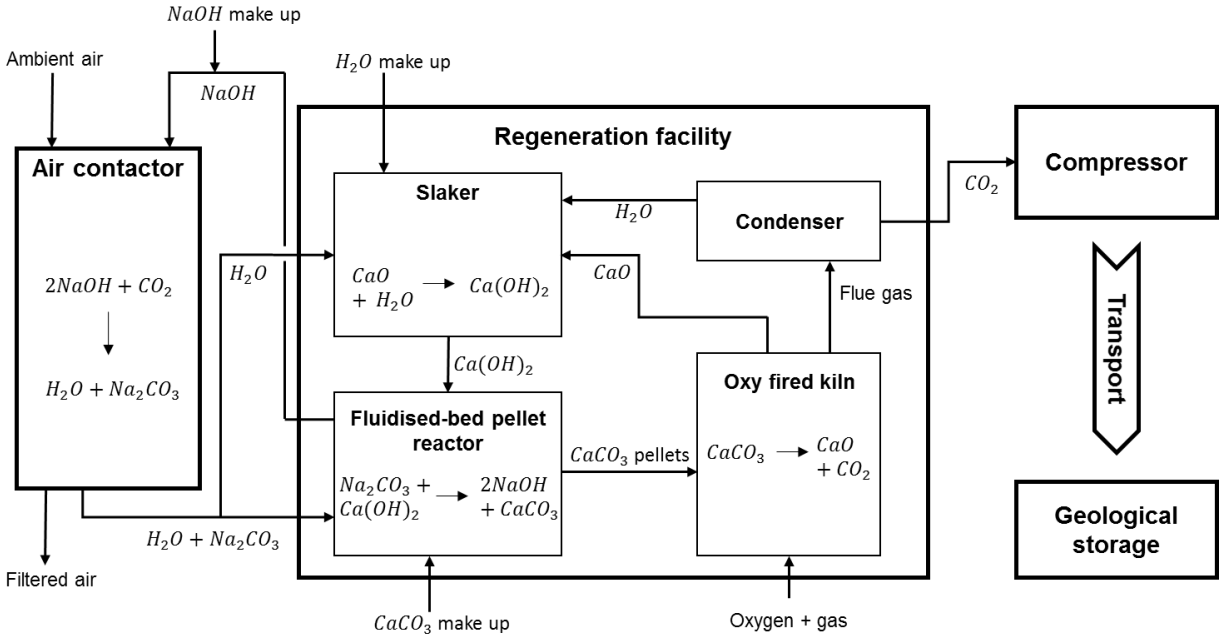

Figure1: Schematic representation of a Direct Air Capture (DAC) facility for the capture and storage of ambient CO_2.

The air contactor units in this analysis are based on a cost-optimized design by Carbon Engineering (Holmes and Keith 2012; base case). Each unit consists of 4 stacked rows of 40 modules with a width and height of 5 m and a depth (d) of 8.6 m. These modules are filled with a PVC packing material over which a strong hydroxide sorbent material is distributed from the top while outside air is blown through the packing material horizontally by fans. The sorbent in this system is a 2 M sodium hydroxide (NaOH) solution. The NaOH solution forms

a thin film on the packing material to create a large surface to volume ratio within the air contactor. The sorbent reacts with CO_2 rich outside air:

$$2NaOH_{(aq)} + CO_{2(g)} \rightarrow H_2O_{(l)} + Na_2CO_{3(aq)} \quad \Delta H° = -109.4\ kJ/mol \qquad \text{Eq. 1}$$

Active fans create a pressure drop within the air contactor to guide outside air through the packing material. The saturated sorbent reaching the bottom of the contactor unit is then pumped to the regeneration facility where the NaOH solution is regenerated and CO_2 is separated. A small scale outdoor prototype of this air contactor was recently tested (Holmes et al. 2013).

The sorbent is regenerated using a 'Pelletized' variant of the Kraft process, as suggested by Holmes et al. (2013) and described as process option B by Baciocchi et al. (2006).The first step in the regeneration process is causticization, where the saturated sorbent (Na_2CO_3) reacts with calcium hydroxide ($Ca(OH)_2$) to regenerate the sorbent and form calcium carbonate ($CaCO_3$):

$$Na_2CO_{3(aq)} + Ca(OH)_{2(s)} \rightarrow 2NaOH_{(aq)} + CaCO_{3(s)} \quad \Delta H° = -5.3\ kJ/mol \qquad \text{Eq. 2}$$

In the pelletized variant of the Kraft process, this step is performed in a fluidised-bed type pellet reactor. The supernatant NaOH solution can be directly pumped back to the air contactor. As the pellets in the reactor grow they sink to the bottom where they are periodically removed from the reactor. The removed pellets are then dried to the atmosphere resulting in calcium carbonate pellets with a solid content of about 90%. The dried pellets are transported to a kiln where they are heated under high temperatures (± 900 °C) to form calcium oxide (CaO, quicklime) and pure CO_2 in the calcination step (reaction 3).

$$CaCO_{3(s)} \rightarrow CaO_{(s)} + CO_{2(g)} \quad \Delta H° = +179.2\ kJ/mol \qquad \text{Eq. 3}$$

In this study we assume that calcination takes place in an oxy fuelled kiln to avoid the production of nitrogen in the produced gas stream and to increase kiln efficiency. The calcination step will thus need an oxygen feed in addition to a fuel feed. The separated CO_2 and the CO_2 generated by heat production in the kiln are mixed into a pure CO_2 stream at a pressure of 1 bar ready for compression. The quicklime produced in reaction 3 is transported to a slaker where it undergoes an exothermic reaction with water, generated in the air contactor (reaction1), to regenerate the calcium hydroxide used during causticization:

$$CaO_{(s)} + H_2O_{(l)} \rightarrow Ca(OH)_{2(s)} \quad \Delta H° = -64.5\ kJ/mol \qquad \text{Eq. 4}$$

The final steps of the DAC system: compression, transport and geological injection are also used in conventional Carbon Capture and Storage (CCS) systems at power plants. CO_2 leaving the kiln is compressed in a four stage compressor to 110 bar for transport trough a natural gas pipeline to an onshore natural gas storage injection site 50 km away. These steps are described by (Koornneef et al. 2008) for a power plant CCS facility with a CO_2 capture rate of 3Mt/yr.

2.2 CO_2 capture rate

While the system described above was designed for an annual capture rate of 1 Mt CO_2 the actual capture rate may deviate from this depending on the obtained mass transfer coefficient (K_L, m/s). The obtained mass transfer rates are expected to lie between 1.1 and 2 * 10^{-3} m/s. We calculated the CO_2 capture rate (r_{CO2}, kg/yr) as (Holmes and Keith 2012):

$$r_{CO_2} = v * \left(1 - e^{-\varepsilon * SSA * d * \frac{K_L}{v}}\right) * t_{op} * A_{unit} * N_{units} \qquad \text{Eq. 5}$$

Where v is the air velocity inside the contactor which was determined by (Holmes and Keith 2012) to be ideally set to 1.6 m/s, d is the air travel distance which is 8.6 m, SSA is the specific packing area of the packing material which is equal to 210 m^2/m^3, ε is the packing efficiency (80%), t_{op} is the yearly operational time in seconds, A_{unit} is the frontal area of one capture unit (4000 m^2) and N_{units} is the total number of air contacting units (10 units).

2.3 Inventory

We included greenhouse gas (GHG) emissions from the materials needed for the construction of the different facilities and GHG emissions from the material, water and energy use during operations. We gathered the necessary inventory data from different sources depending on the system component, as further explained below.

2.3.1 Construction

The infrastructure of the air contactor was based on the description provided by Holmes and Keith (2012) for a contactor unit designed to have a capture rate of 0.1 Mt CO_2/yr and an assumed average life time of 20 years. The packing volume and materials, the inlet louver materials, the drift eliminator materials and the number and type of fans were readily taken from the descriptions (Holmes and Keith 2012). For the foundation and casing of the air contactor we assumed that reinforced concrete was used with a foundation depth of 3 m and a wall thickness of 20 cm. The number of spray nozzles was estimated from the number of nozzles per m^2 used in the prototype (Holmes et al. 2013) and the basal area of the proposed contactor. For the cross flow nozzles we estimated a total of 100 grams of polypropylene was used per nozzle. Furthermore, we estimated a total of 500 tonnes of low-alloyed steel is needed for use in fluid pumps, fluid pipes, shut-off valves and other small equipment per contactor unit. The DAC system in this study consisted of 10 of the described contactor units.

The necessary infrastructure for the regeneration facility has not been described as far as we know. Therefore, we excluded GHG emissions related to the construction of the regeneration infrastructure in our analysis. The inventory for the infrastructure of the compressor and injection facility and the pipelines for transportation were based on a detailed inventory for a power plant CCS facility with a capacity of 3MtCO_2 captured per year (Koornneef et al. 2008). LCI data for the production process of the necessary infrastructure materials are taken from the Ecoinvent database (version 3.3; Werner et al. 2016).

2.3.2 Energy requirements

Annual mechanical energy requirements for the fans in the air contactor (E_{fans}, J/yr) were calculated from the yearly operational time (t_{op}, s), air velocity (v, m/s), air travel distance (d, m), and fan efficiency (η_{fans}):

$$E_{fans} = d * \frac{7.4v^{3.14}}{\eta_{fans}} * t_{op} * A_{unit} * N_{units} \quad \text{Eq. 6}$$

Mechanical energy requirements for the sorbent pumps are calculated using a sorbent flow rate (f) of 0.41 l/(m^2s) which was determined to be an optimal trade of between capture rate and energy costs in the outdoor prototype of the air contactor by Carbon Engineering (Holmes et al. 2013). For a contactor with a total of 10 units consisting of 4 stacked rows of 40 modules the annual energy requirements for the sorbent pumps (E_{pump}, J/yr) can then be calculated as:

$$E_{pumps} = \frac{w*d*\rho_{Sorbent}*f*g*10h*40}{\eta_{pumps}} * t_{op} * N_{units} \quad \text{Eq. 7}$$

Where w is the width of a module (5 m), d is the depth of a module (8.6 m), $\rho_{Sorbent}$ is the density of the 2M NaOH sorbent (1.08 kg/l), g is 9.81 m/s^{2}and h the height of a module (5 m). Energy requirements for the fans and sorbent pumps were divided by the annual capture rate as calculated in section 2.2.1 to obtain energy requirements per ton CO_2 captured.

The energy requirements for the regeneration process are based on an earlier study by Baciocchi et al. (2006) including a pellet reactor for causticization and an oxy-fired calcination kiln with an oxygen feed supplied by a Pressure Swing Adsorption (PSA) process. To calculate the mechanical energy requirements for the work done by the pellet reactor and slaker we assumed an electromotor efficiency of 95% for the slaker and a pump efficiency to 75% (equal to the sorbent circulation pumps) for the pellet reactor, leading to an energy requirement of 42 kWh/tCO2. Additionally, the PSA system generating the oxygen stream to the kiln has an energy requirement of 116 kWh/tCO_2 (Baciocchi et al. 2006).

Heat demand (4470 MJ/tCO_2) in the kiln is met by combustion of natural gas. CO_2 emissions generated during heat production in the calcination kiln are mixed with the CO_2 separated from the sorbent. We assumed that the heat released during the cooling of the gas and CaO streams out of the kiln can be captured and used with an efficiency of 90%. This leads to a heat demand of 1812 MJ/tCO_2 additional to the heat demand in the calcination kiln which is again assumed to be met by on site combustion of natural gas.

Energy requirements for the compression and geological injection of CO_2 were taken from Koornneef et al. (2008). Mechanical energy requirements in the baseline scenario were assumed to be met by a natural gas combined cycle power plant with a carbon footprint of 0.50 kg CO_2-eq./kWh generated calculated for a 100 year time horizon and with Global Warming Potentials (GWP) from Stocker (2014) (Hauck et al. 2014; S5). An overview of all energy considerations in the system is given in Table 1.

2.3.3 Water loss

Water loss in the air contactor was calculated from the CO_2 capture fraction (ΔP_{CO2})the vapour pressure of water at ambient temperature ($P_v(T)$), the relative humidity of the ambient air (R_H) and the saturation of water in the sorbent (S) (Socolow et al. 2011):

$$r_{H_2O/CO_2} = \frac{P_v(T)*S - P_v(T)*R_H}{\Delta P_{CO_2}} \quad \text{Eq. 8}$$

For a 2M NaOH solution the saturation constant can be estimated as the mole fraction of water in the sorbent (~97%). In the baseline scenario we set the relative humidity to 65% and the temperature to 13 °C leading to a loss of about 4.6 mol H_2O per mol CO_2.

2.3.4 Sorbent and lime requirements

Operation of the DAC system requires the input of chemicals and water. While the reaction cycles used for regeneration are closed in theory (figure 1), recycling efficiencies never reach 100% and chemical losses must be accounted for. In our analysis we account for these losses in the amount of NaOH and $CaCO_3$ needed. Total requirements ($x_{i,total}$) of each compound i were calculated using the recycling efficiency (0 < R < 1) and equation 2.

$$x_{i,total} = x_{i,base}\ (1 - R_i) \quad \text{Eq. 9}$$

Where $x_{i,base}$ is the amount of compound i in moles required to capture 1 mole of CO_2 when no recovery of the compound is possible, i.e. a one pass process

Using the specifications arriving from the cost optimalization study done by Holmes and Keith (2012) we assume that partially loaded sorbent leaving the air contactor is recirculated back to air contactor so that the sorbent entering the regeneration process has a CO_2 loading of

about 20 g CO2 per kg sorbent. Based on a 2M NaOH solution this then leads to a one pass requirement of about 4.8 mol NaOH per mol CO_2 captured. However, because the system we analyse here uses a recycling step for the sorbent, the partially loaded sorbent leaving the air contactor is regenerated during causticization. For this study we based the regeneration efficiency of NaOH on the mass balances developed by Baciocchi et al. (2006) for this regeneration process. We calculated the efficiency as the amount of Na^+ leaving the pellet reactor in the regenerated sorbent stream divided by the amount of Na^+ fed into the pellet reactor, leading to a recycling efficiency of 99.9% (Baciocchi et al. 2006). Using Eq. 9 and the earlier calculated amount of 4.8 mol NaOH per mol CO_2 captured as a base value gives actual NaOH requirement of 0.003 mol NaOH per mol CO_2.

Losses in the calcination kiln and slaker are accounted for in the total amount of $CaCO_3$ needed. Similar to the approach for NaOH, we calculated the efficiency of $CaCO_3$ recycling from the mass balances developed by Baciocchi et al. (2006) resulting an efficiency of 90.9%. Using stoichiometric amounts of $CaCO_3$ as a base (eq. 3), this results in a total $CaCO_3$ requirement of about 0.1 mol $CaCO_3$ per mol CO_2.

2.4 Carbon capture efficiency

We calculated the carbon efficiency of the DAC system as:

$$E_c = \frac{CO_{2,cap} - GHG_{lc}}{CO_{2,cap}} * 100 \qquad \text{Eq. 10}$$

where CO_{2cap} is the total amount of kg CO_2 stored over the lifetime of the system and GHG_{lc} is the total amount of greenhouse gasses emitted over the whole life cycle of the system in kg CO_2 equivalents. GHG emissions related to the construction, chemical and water requirements were calculated with the Ecoinvent database (Version 3.3; Wernet et al. 2016). Global Warming Potentials (for a 100 year time horizon) to sum up GHG emissions to kg CO_2-equivalents were taken from the most recent IPCC-report (Stocker 2014). When $E_c<0$, more carbon is emitted over the lifetime of the system than is captured, while a $E_c>$ 0indicates that more CO_2 is stored than emitted over the life cycle.

2.5 Sensitivity analysis

For various system parameters, i.e.: the mass transfer coefficient and the lifetime of the facility, various ranges can be found in literature. To estimate the influence of these uncertainties, we determined an optimistic and pessimistic value based on the reported ranges next to the baseline value (Table 1). Other parameters, i.e.: the ambient air temperature, the relative humidity, the electricity source and the amount of heat recovered during the regeneration, depend on choices made during the design of the facility. For these parameters, we determined alternative options for the optimistic and pessimistic scenario. For the electricity source, we added to the optimistic scenario the use of solar power as this is an often mentioned possibility in these systems (House et al. 2011, McGlashan et al. 2012). Additionally, in the pessimistic scenario, we included a scenario where the electricity demand is met by an average US coal plant (Steinmann et al. 2014). GHG life cycle emissions of the alternative energy sources are given in Table 1. We also investigated two alternative regeneration heat demand scenarios: one in which heat released during the condensation step is recovered and reused (optimistic) and one in which none of the heat released (including gas and CaO streams leaving the kiln) is used (pessimistic) (Baciocchi et al. 2006). Lastly, the geographical placement of the facility can have a large impact on the water use of the system, to account for this, we set R_H=82% with T=9.5°C in the best case scenario, corresponding to the yearly average for the Netherlands, and in the worst case scenario we set R_H=38% with T=21 °C, corresponding to the yearly average in Tucson, Arizona. We performed a sensitivity analysis by calculating the E_c for the optimal combination

and the pessimistic combination of the abovementioned parameter values. We also obtained for each of the above described parameters the influence on the E_c.

3. Results

Using the baseline inventory data, we obtained a positive E_c of 62%. Figure 2 shows the amount of CO_2-eq. emitted per tonne of CO_2 captured from the different life cycle components of the DAC system. The majority of the GHG emissions from the DAC system are caused by the energy demand, approximately 65% for electric energy demand and 32% for heat demand in the baseline scenario.Water use, sorbent use and $CaCO_3$ use both contribute less than 1% to the total greenhouse gas emissions of the DAC system.

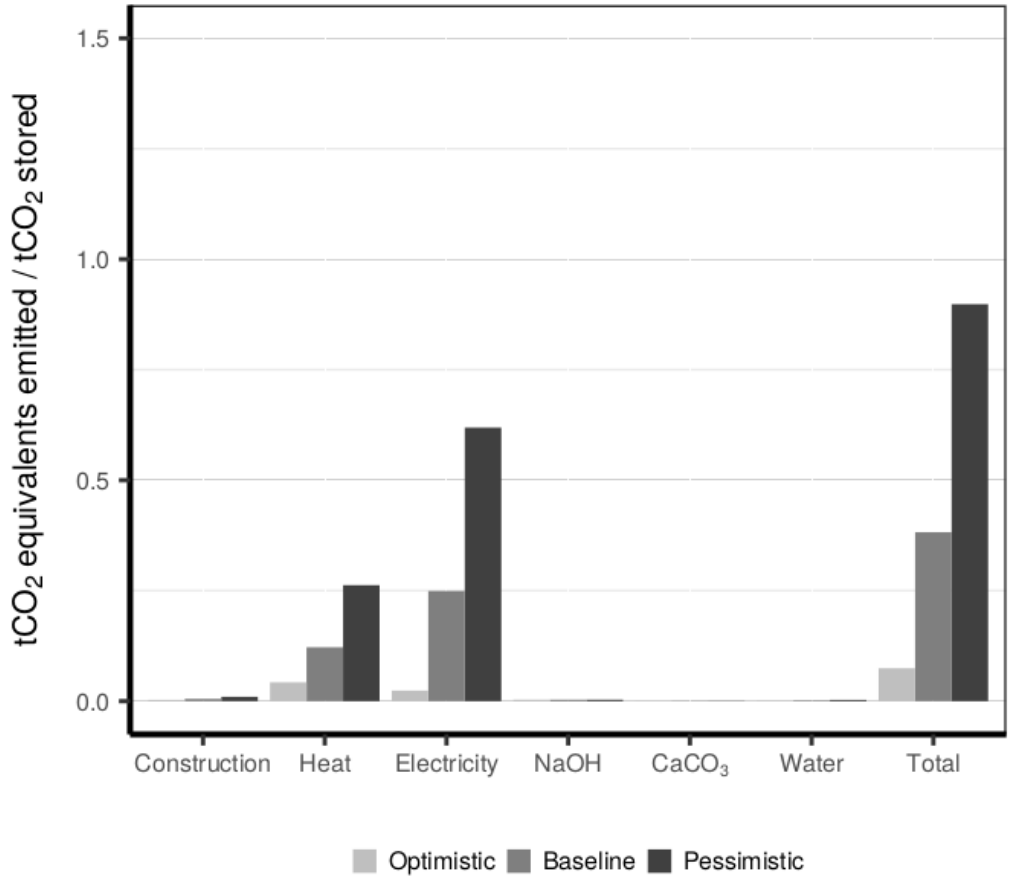

Figure2: Life cycle greenhouse gas emissions emitted per amount of carbon stored from various components of a Direct Air Capture (DAC) facility based on strong hydroxide sorbents utilizing the Kraft process for sorbent recovery. Carbon emissions were calculated in three scenarios: optimistic, baseline and pessimistic, in which mass transfer coefficient, temperature, relative humidity, heat recovery during the regeneration process, fan and pump efficiencies, lifetime and carbon footprint of the electricity source were set to their optimal, basis and worst values respectively.

For the optimistic and pessimistic scenarios, we found a E_c of 92% and 10%, respectively. The results of the sensitivity analysis are shown in figure 3. Switching to a renewable energy source to meet the energy demand by the fans, pumps, compressor and injection facility has the largest potential to increase the E_c. Using solar power to generate the required electricity may improve the E_c from 62% to 84% using the baseline scenario for all other parameters. Recovery of the heat released during condensation may also improve the E_c with about 8%.

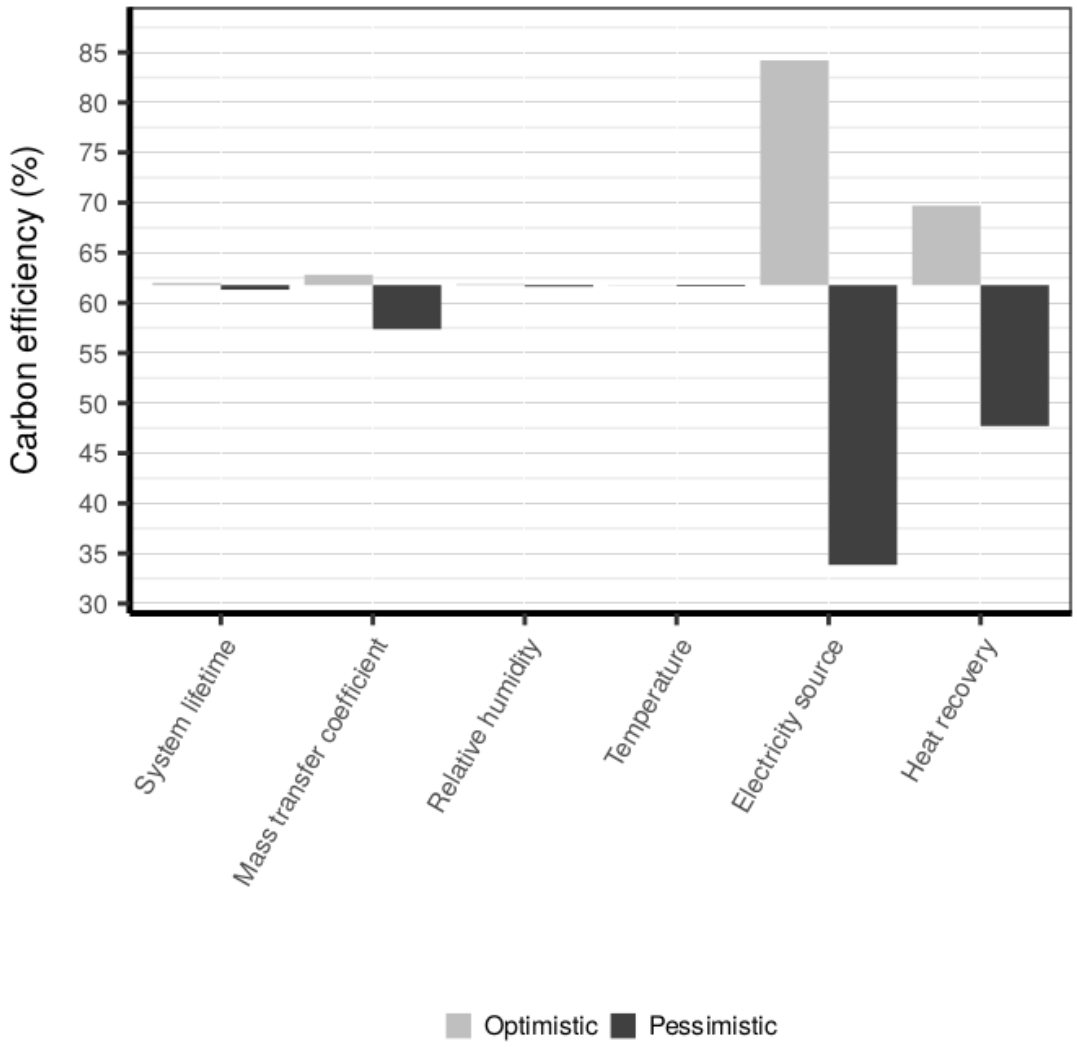

Figure3: Change in life cycle carbon efficiency of a Direct Air Capture (DAC) system, using a 2M NaOH solution as a sorbent and utilizing the pelletized variant of the Kraft process for sorbent regeneration, with respect to the baseline due to optimistic and pessimistic parameterizations.

Other parameters in our sensitivity analysis only have a minor effect on the E_c.

4. Discussion

The main goal of this study was to assess the life cycle carbon efficiency (E_c) of direct capture of CO_2 from ambient air. The DAC system analysed in this study was a system based on a strong hydroxide sorbent which was regenerated using the pelletized variant of the Kraft process. In our baseline scenario the system has a high positive E_c indicating that DAC can effectively contribute to climate change mitigation. Even in the worst case scenario, the system still effectively captures CO_2.

Several studies before us have analysed the heat and energy requirements of hydroxide based DAC systems (House et al. 2011, Goeppert et al. 2012, Sanz-Pérez et al. 2016). From these studies it, however, remained unclear what the potential carbon benefits of the DAC system are because heat and energy demands differed substantially between studies. Secondly, these studies neglected the contribution of other system characteristics, such as the use of sorbent and GHG emissions from infrastructure. In our study, we showed that, emissions from these sources do not significantly contribute to the overall carbon efficiency of the system.

GHG emissions due to the electricity demand have the largest impact on the life cycle E_c. While we do not expect that the total mechanical demand of the system can be significantly decreased, the electricity source can play a major role in the carbon efficiency (figure 3). When assuming an industrial PV installation is used to meet the electricity demand (optimistic scenario), the emissions from the mechanical energy demand become negligible compared to the other system emissions. Making sure that mechanical energy requirements are provided by a renewable energy source is an important step in further improving the carbon efficiency of this system. Some authors have also cited the use of KOH instead of NaOH as a possible option because it has faster reaction kinetics with CO_2 (Bandi et al. 1995, Holmes et al. 2013, Sanz-Pérez et al. 2016). This will decrease the amount of energy needed to circulate the sorbent throught the contactor thus lowering the mechanical energy demand of the system. However, according to Ecoinvent (version 3.3; Wernet et al. 2016) the life cycle GHG emissions of KOH (2.2 kg CO_2 eq/kg KOH) is higher than that of NaOH (1.4 kg CO_2 eq/kg NaOH). Switching from NaOH to KOH might therefore not lead to an increased carbon efficiency if the increase in CO_2 capture rate is not sufficient to offset the higher life cycle GHG emissions of KOH production.

The large energy requirements of the regeneration process, and especially the energy requirements by the calcination kiln are generally seen as the limiting factor in NaOH based DAC systems. We showed that an increase in system carbon efficiency of 8% can be achieved when the heat released during condensation is recovered and used in the $CaCO_3$ heating and drying steps. However, Baciocchi et al. (2006) pointed out that this is low-grade heat that is difficult to recover. This improvement may thus not be feasible in practice. Alternative options for reducing the heat demand have been suggested, such as using the heat that is released during slaking in the calcination step (Zeman 2007). However, this option has also been criticized as impractical (Baciocchi et al. 2006, Stolaroff 2006). Alternatively, Mahmoudkhani and Keith (2009) proposed to use causticization by titanium dioxide (TiO_2) instead of the Kraft process for sorbent regeneration. They proposed to regenerate the NaOH using direct causticiation with TiO_2 (Mahmoudkhani and Keith 2009). Using TiO_2 the regeneration of NaOH was estimated to require 50% less heat. However, the greenhouse gas emissions (in kg CO_2 eq.) related to the production of 1 kg of TiO_2 are about 1800 times higher than those for 1 kg of $CaCO_3$ (ecoinvent version 3.3; Wernet et al. 2016). TiO_2 regeneration efficiencies well over 99% would be necessary for this to be a feasible option. Furthermore, although the calcination step is a large contributor to the overall energy requirements of the system, it only has a minor contribution to the systems carbon efficiency as all CO_2 emitted by combustion in the kiln is mixed and stored with the captured CO_2.

The contribution of water loss to the systems carbon efficiency was found to be negligible. However, many studies on the use of strong hydroxide solutions for carbon capture have implicated the use of water by the system as a possible bottleneck with values ranging from 1 to 30 mol H_2O per mol CO_2 (Zeman 2007, Stolaroff et al. 2008). This range is much larger than the range calculated in our assessments using eq. 8. Using the high end of this range (30 mol H_2O per mol CO_2) however only decreases the carbon efficiency with 1%. Even higher values for evaporative water losses have been reported by Zeman (2008) when including the regeneration process. However, water loss by evaporation during the regeneration process is minimized by the use of the fluidized bed pellet reactor instead of a more common precipitator. While we found that water loss only had a minor impact on the carbon efficiency, its impact on other impact categories may, however, also be important to study. To fully assess whether DAC systems are a feasible climate change mitigation option, these environmental impacts need to be quantified in future research. However, because direct air capture systems are not bound to point emission sources, they can be located in areas with sufficient water and a low-carbon electricity supply.

Compared to the impact of the energy needed during operations of the DAC system, infrastructure only had a minor contribution. Data availability to perform a full inventory analysis of the infrastructure was one of the major limitations of this study. Because we omitted the infrastructure for the regeneration facility, the carbon efficiency of the DAC system will be lower than reported here. However, the contribution of infrastructure to the life cycle GHG emissions of a technology is generally low in systems with relatively high energy and chemical requirements (Frischknecht et al. 2007). This is also the situation for the DAC system which is readily shown by the small contribution of the infrastructure of the DAC facility itself in the current analysis (<1%).

It is important to further note that the data used for our study is derived from systems with varying technological readiness level (TRL; Gavankar et al. 2015). The air contactor described in this study has already been tested using an outdoor prototype. However, the data for the regeneration process is based on systems with widely varying TRL. For example, the slaker and the pellet reactor have been used on a large scale while the characteristics of the oxygen fuelled calcinations kiln were based only on a formulation of a kiln that was not yet developed (Baciocchi et al. 2006). Furthermore, data for the compressor and transportations pipelines have been based on level 9 technologies (Koornneef et al. 2008). An overview of the TRL of the systems underlying the data used in this study can be found in the SI.

Gavankar et al. (2015) noted that the emissions can drastically decrease with increasing TRL because of recycling of the feed stock. We already accounted for recycling in our analysis by including the estimations of regeneration efficiency and we thus do not expect emissions to significantly decrease. Furthermore, this technology has recently been developed and tested on a prototype level by Carbon Engineering (Keith et al. 2018). Using this prototype, Keith et al. (2018) calculated a slightly lower overall heat demand of 5.25 GJ/tCO2, compared to 6.28 GJ/tCO2 estimated in our baseline. However, as all CO_2 generated in the kiln is captured, this would lead to an increase of the carbon efficiency of only 6%. Furthermore, the carbon efficiency found with the optimal parameterization in this study is comparable to the preliminary LCA results reported for this prototype by Keith et al. (2018). These results indicate that the system analysed here may be an effective climate change option, especially when renewable energy sources are used for electricity generation.

5. Conclusions

In this study we calculated the carbon efficiency of a Direct Air Capture system over its full life cycle. As DAC systems are still relatively new, full assessment of the greenhouse gas emissions during the system lifetime was challenging. In this study, we analysed a DAC system based on strong hydroxide sorbents as a climate change mitigation option. Our results showed that this system has a positive carbon efficiency, meaning that it can effectively capture and store CO_2 from ambient air. Furthermore, we showed that the most important emissions sources of the system are the energy demand. The largest improvements in the carbon efficiency of this system can be achieved by using renewable energy sources to meet the system's mechanical energy demand, which is not only the largest contribution to carbon efficiency but also easiest achieved from a technical point of view. In its current form, it is concluded that the DAC system is an effective climate change mitigation option based on its full carbon balance.

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Supplementary materials

Data S1: Spreadsheet containing the energy, chemical and water use of the system and the full inventory.

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551 # Tables

552 *Table 1: Input parameters for the life cycle GHG analysis of a Direct Air Capture system based on a 2 M NaOH sorbent*
553 *utilizing the pelletized Kraft process for sorbent regeneration.*

	Baseline	Optimistic	Pessimistic
System characteristics			
Lifetime (yr)	20	40	10
Percentage year operations (%)[a]	85	-	-
Mass transfer coefficient (K_L, mm s^{-1})[a]	1.5	2.0	0.8
Air velocity (v, m s^{-1})[a]	1.6	-	-
Sorbent flow (f, L m^{-2} s^{-1})[b]	0.41		
Ambient CO2 concentrations (ppm)	400	-	-
Packing efficiency (ε, %)[a]	80	-	-
Specific packing area (SSA, m^2 m^{-3})[a]	210	-	-
Air travel distance (d, m)[a]	8.6	-	-
Fan efficiency (η_{fan}, %)	56	-	-
Liquid pumps efficiency (η_{pump},%)	75	-	-
Relative humidity (R_H, %)	65	82	38
Temperature (T, °C)	13	9.4	21
NaOH recycling efficiency (%)[c]	99.9	-	-
$CaCO_3$ recycling efficiency (%)[c]	90.9	-	-
Efficiency heat transfer (%)	90	-	-
Carbon footprint electrical energy source (kg CO_2 eq. kWh^{-1})	0.50[d]	0.05[e]	1.06[f]
Infrastructure inventory			
Air contactor[b]			
PVC (t)	$14 * 10^3$	$14 * 10^3$	$14 * 10^3$
Concrete foundation (m^3)	$1.1 * 10^5$	$1.1 * 10^5$	$1.1 * 10^5$
Concrete (m^3)	$70 * 10^2$	$70 * 10^2$	$70 * 10^2$
Polypropylene (t)	15	15	15
Stainless steel (t)	32	32	32
Grass-fibre reinforced polyurethane (t)	70	70	70
Low-alloyed steel (t)	$50 * 10^2$	$50 * 10^2$	$50 * 10^2$
Compressor, transport and storage			
See Koornneef et al. (2008)			
Energy balance			
Air contactor			
Electricity (kWh $tonne^{-1}$ CO_2 captured)	193	172	278
Regeneration			
Electricity O_2 purification (kWh $tonne^{-1}$ CO_2 captured)	116	-	-
Electricity Slaker (kWh $tonne^{-1}$ CO_2 captured)	1.43	-	-
Electricity pellet reactor (kWh $tonne^{-1}$ CO_2 captured)	40.7	-	-
Heat CaCO3 drying and air heating(MJ $tonne^{-1}$ CO_2 captured)	3900	-	-
Heat CaO cooling (MJ $tonne^{-1}$ CO_2 captured)	- 864	-	0
Heat flue gas cooling (MJ $tonne^{-1}$ CO_2 captured)	- 1224	-	0
Heat water condensation (MJ $tonne^{-1}$ CO_2 captured)	0	- 1179	-
Heat calcination (MJ $tonne^{-1}$ CO_2 captured)	4470	-	-
Compression			
Electricity (kWh $tonne^{-1}$ CO_2 compressed)[g]	111	111	111
Geological injection			
Electricity (kWh $tonne^{-1}$ CO_2 injected)[g]	7	7	7

[a] Values adapted from the cost optimized air contactor design by (Holmes and Keith 2012).
[b] Based on an optimal flow rate found in the prototype air contactor by (Holmes et al. 2013).
[c] Adapted from (Baciocchi et al. 2006).
[d] Carbon footprint for an average natural gas combined cycle power plant in the US calculated using a time horizon of 100 years and recently updated GWPs (Hauck et al. 2014).
[e] Carbon footprint for an average solar photovoltaic power plant (Nugent and Sovacool 2014)
[f] Median carbon footprint found for US coal fired power plants based on a 100 year time horizon (Steinmann et al. 2014)
[g] Values taken from (Koornneef et al. (2008))

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